PATENT COOPERATION TF - ATY

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Date of mailing (day/month/year) 30 August 2000 (30.08.00)	in its capacity as elected Office
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Applicant	
CHOI, Ki-Seung et al	
The designated Office is hereby notified of its election made. X in the demand filed with the International Preliminary 13 June 2000 in a notice effecting later election filed with the International Preliminary	(13.06.00)
2. The election X was was not was not made before the expiration of 19 months from the priority of Rule 32.2(b).	date or, where Rule 32 applies, within the time limit under

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BIOCIDE COMPOSITION AND STERILIZATION METHOD USING THE SAME

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This application is based on application No. 98-49095 filed in the Korean Industrial Property Office on November 16, 1998, the contents of which are incorporated hereinto by reference.

BACKGROUND OF THE INVENTION

(a) Field of the Invention

The present invention relates to a biocide composition, more particularly to a biocide composition having synergistic effects by comprising 3-isothiazolone and polyhexamethyleneguanidine phosphate and a method for sterilizing microorganisms, fungi, etc. using the same.

(b) Description of the Related Art

Microorganisms, bacteria, mold, algae, etc. grow in industrial water such as cooling water, wastewater, emulsifiers used in the textile industry, etc. and have a deteriorating effective on the operations of industrial processes. Such microorganisms propagate using organisms contained in the industrial water as a nutrient source and secrete polysaccharides. Varieties of organic and inorganic substances combined with these secreted polysaccharides and form 20 viscous lumps or masses also called slime. Organic materials such as cellulose, hemicellulose, and the fibrin of white water in paper industry in particular provide to have sufficient nutrient sources for such microorganisms. The slime formed at areas of low fluid flow in a paper processing process results in both direct and indirect losses, such as manufacturing time loss, equipment 25 efficiency deterioration, etc. due to the deterioration of pulp quality, etc. Furthermore, the growth of microorganisms at such places like a cooling water facility, where much water is contained or recirculated causes a fouling phenomena. This deteriorates heat transfer efficiency in an industrial cooling tower, as well as corrodes metal or erodes wooden parts..

Bacteria such as taloblastic prokaryotes can propagate by decomposing

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various types of organisms, some of which are the sources of the microbilogically induced corrosion, by secreting and discharging polysaccharides forming a biofilm. Fungi such aseukaryote can also propagate by decomposing various types of organisms like bacteria, and some types of fungi secreting cellulase are the sources for discoloration and decomposition of wooden parts by way of degrading the fibroid materials of wooden parts of a cooling tower, etc.

Algae, such as eukaryote, can propagate by photosynthesizing under an environment with light, air, and a small amount of organisms, and carbohydrate formed from algae is used as a nutrient source for other microorganisms such as bacteria and fungi, thereby accelerating the fouling phenomena. Algal fouling due to algae propagation is intensified at places that are exposed to sunlight, particularly in cooling water facilities, swimming pools, etc. This phenomena results in clogging which blocks water pipes, as well as the deterioration of heat transfer efficiency, oxidization of metal surfaces by the generation of oxygen, and the promotion of corrosion at a holes on metallic surfaces through a partial galvanic reaction when the organism dies.

Various biocides are being developed in order to kill such microorganisms, fungi, algae, etc. or to prevent their surface adhesion to metal, etc. These biocides are generally divided into oxidant biocides and non-oxidant biocides. The oxidant biocides that are mainly used are halogen compounds such as chlorine, bromine, etc. that are popular due to their economic advantage of strong oxidation capability and low price. However, they can cause erosion of the wooden parts of cooling towers and metallic decomposition, and their sterilizing efficiency tends to deteriorate as they can be easily being discharged into atmosphere. Furthermore, their practical effectiveness is poor due their peculiar way of first reacting with secreted polysaccharides before they react with microorganisms that are the actual sources of biofilm formation.

Non-oxidant biocides which overcome these disadvantages include 3isothiazolone, quaternary ammonium salt, formaldehyde emission compound, glutaraldehyde, etc. and are mainly used separately. Although 3-isothiazolone, which is disclosed in United States Patent Nos. 3,761,488, 4,105,431, 4,279,762, etc., has a high sterilizing effect and wide antibiotic spectrum, it has a

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disadvantage in that its immediate instantaneous sterilizing effects are low. Furthermore, Korean Patent Application No. 89-20381 discloses an antiseptic composition useful in preventing circulation water putrefaction wherein a biocides 5-chloro-2-methyl-4-isothiazolone-3-on and 2-methyl-4-isothiazolone-3-on are mixed in a ratio of about 3:1 and wherein this antiseptic composition further comprises didecyldimethylammoniumchloride. United States Patent No. 4,379,137 discloses a method for improving sterilizing capability by mixing polymer quaternary ammonium salt and 3-isothiazolone. However, since these mixtures emit corrosive materials, i.e., halogen compounds such as fluorine, chlorine, etc., it is difficult for them to be applied where metals susceptible to corrosion are used, such as carbon steel, cast iron, stainless steel, copper, etc.

Additionally, Korean Patent Application No. 97-80170 discloses a process that can be applied even when metals susceptible to corrosion are used since the disclosed process does not emit a halogen compound. Another water soluble biocide composition having synergistic effects comprising 3-isothiazolone, which has high sterilizing effect on quarternary ammonium phosphate and microorganisms, is disclosed in Korean Patent Application No. 97-46517 as a biocide having superior properties of immediate sterilizing effects on microorganisms, durability, anticorrosiveness, etc. However, there is a problem in applying this invention to various industrial fields due to the issues such as bubbling or foaming, etc. when quaternary ammonium is added to provide immediate effectiveness to the 3-isothiazolone.

Furthermore, while polyhexamethyleneguanidine phosphate has immediate effectiveness, and is used in the effective and wide control of microorganisms in various industrial fields, including water treatment, as well as exhibiting low foaming properties, it has a disadvantage of not having a wide antibiotic spectrum by itself.

SUMMARY OF THE INVENTION

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It is an object of the present invention to provide a biocide composition which can be used in the process where metals susceptible to corrosion are used, such as carbon steel, cast iron, stainless steel, copper, etc., as well as which has high sterilizing capability, a wide antibiotic spectrum, and superior

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antiseptic effects.

It is an other object of the present invention to provide a biocide composition which can not only be applied to various industrial processes on account of its low foaming features, but also which has a high sterilizing capability even when a small amount is used.

The present invention provides a biocide composition comprising 3-isothiazolone of the following General Formula 1 and polyhexamethyleneguanidine phosphate of the following General Formula 2 in order to accomplish the above objects:

[General Formula 1]

$$H_3C-N$$

[General Formula 2]

$$\begin{array}{c} \text{NH} \\ \parallel \\ -\{(\text{CH}_2)_6 - \text{NH} - \text{C} - \text{NH} - \frac{1}{m} \cdot \text{nH}_3 \text{PO}_4 \end{array}$$

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where R is hydrogen or chlorine in the above General Formula 1 in the above General Formula 1, m is integer from 4 to 7, and n is an integer from 1 to 14 in the above General Formula 2. The present invention further provides a sterilizing method which kills or restrains the growth of bacteria, fungi or algae by inputting the above biocide composition into a media that is contaminated by

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bacteria, fungi, and algae.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

In the following detailed description, only the preferred embodiments of the invention have been shown and described, simply by way of illustration of the best mode contemplated by the inventor(s) of carrying out the invention. As will be realized, the invention is capable of modification in various obvious respects, all without departing from the invention. Accordingly, the description is to be regarded as illustrative in nature, and not restrictive.

The present invention is described in detail below.

The present invention provides an antiseptic and/or biocide composition comprising 3-isothiazolone of the below General Formula 1 and polyhexamethyleneguanidine phosphate of the below General Formula 2:

[General Formula 1]

[General Formula 2]

$$\begin{array}{c} \text{NH} \\ \parallel \\ -\{(\text{CH}_2)_6 - \text{NH} - \text{C} - \text{NH} - \frac{1}{m} \cdot \text{nH}_3 \text{PO}_4 \end{array}$$

where R is hydrogen or chlorine in the above General Formula 1 in the above General Formula 1, m is integer from 4 to 7, and n is an integer from 1 to 14 in the above General Formula 2.

In the above General Formulae, 3-isothiazolone is preferably 2-methyl-4-isothiazolone-3-on having R group of hydrogen or 5-chloro-2-methyl-4-

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isothiazolone-3-on having R group of chlorine, and more preferably a mixture in which 2-methyl-4-isothiazolone-3-on and 5-chloro-2-methyl-4-isothiazolone-3-on are mixed in a weight ratio of 1:20 to 20:1. If the above ratio of 3-isothiazolone compound is out of the range of 1:20 to 20:1, the sterilizing effects are deteriorated.

The mixing ratio of the above 3-isothiazolone and polyhexamethyleneguanidine phosphate is preferably a weight ratio of 1:1 to 1:65, more preferably 1:1 to 1:4. If the above mixing ratio of 3-isothiazolone and polyhexamethyleneguanidine phosphate is out of the above range, the synergistic effects by the mixture of two biocides are deteriorated or not seen.

If the present invention of a mixture of 3-isothiazolone and polyhexamethyleneguanidine phosphate are used as a biocide composition, 3-isothiazolone compensates for the disadvantage in the low sterilizing effects of polyhexamethyleneguanidine phosphate on fungi and some bacteria, while polyhexamethyleneguanidine phosphate compensates for the disadvantage of 3-isothiazolone has in its low initial and immediate sterilizing capability and effectiveness. Synergistic effects are seen in that the sterilizing action of a mixed biocide becomes greater than the sum of the sterilizing action that each component compound has. That is, the present invention of a composition can restrain microorganisms more widely and effectively by mixing the two types of compounds having different sterilizing mechanisms, and obtains further effects such as the lowering of an emergence frequency of resistant strains more than in the case of using a single component compound.

A biocide composition of the present invention is preferably used in an aqueous solution and more preferably used by putting 5 to 1,000 parts per million (ppm) of it into cooling water, etc. in which the microorganisms exist, although the amount used has no limit as long as it achieves the targetted sterilizing effects. A biocide composition of the present invention can be used in controlling microorganisms widely and effectively in the various industrial fields such as in pulp and paper plants, cooling towers, as a disinfectant, etc.. In particular, it can be used by adding it to the cooling water of an industrial process, to disinfectant, paint, latex antiseptic, cosmetic additives, additives for emulsion products such as shampoo, etc., slime control chemicals for textile

weaving, paper slime control agent, and antiseptics for leather goods, metal processing oil, etc.

COMPARATIVE EXAMPLES for helping in gaining in an understanding
of the present invention and preferable EXAMPLES are described below.

[EXAMPLES 1-6]

3-isothiazolone of and effects sterilizing The polyhexamethyleneguanidine phosphate on a mixed solution of 8 types of 10 bacteria (Enterobacter aerogens ATCC 13048, Escherichia coli ATCC 11229, Micrococcus luteus ATCC 9341, Pseudomonas aeruginosa ATCC 15442, Klebsiella pneumoniae ATCC 1560, Staphylococcus epidermis ATCC 155, Staphylococcus aureus ATCC 6538, and Bacillus subtilis ATCC 6984) were tested using Tryptic Soy Broth manufactured by Difco Corporation according to 15 the two fold dilution method. The minimum inhibiting concentration of each biocideand mixtures thereof were determined by observation with the naked eye of the degree of muddiness of the lowest concentration in which growth was deteriorated after culturing a culture fluid in which biocides and microorganisms are added at 30 degrees centigrade for 3 days.

It was determined that a synergistic effect of a biocide was seen when the sum of QA/Qa and QB/Qb was less than 1, as in the following EQUATION according to the method published in the paper of Kull, F. C. et al (Appl. Microbiol. 9:53 8~544 (1961)), and these results are represented in Table 1:

Synergistic Index (SI) = (QA/Qa) + (QB/Qb)

where Qa and Qb are MIC values (ppm) of a single compound A and single compound B, respectively, and QA and QB are MIC values (ppm) of the compounds A and B, respectively, out of each mixture.

[Table 1]

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	Qa	Qb	QA	QB	QA/Qa	QB/Qb	SI
EXAMPLE	18.8	312.5	9.4	9.8	0.5	0.03	0.53
EXAMPLE 2	18.8	312.5	9.4	19.5	0.5	0.06	0.56
EXAMPLE	18.8	312.5	9.4	39.1	0.5	0.13	0.63
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EXAMPLE	18.8	312.5	9.4	78.1	0.5	0.25	0.75
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EXAMPLE 5	18.8	312.5	9.4	156.3	0.5	0.50	1.0
EXAMPLE 6	18.8	312.5	2.4	156.3	0.13	0.50	0.63

Qa: MIC value (ppm) on the mixed strain of a single 3-isothiazolone;

Qb : MIC value (ppm) on the mixed strain of a single polyhexamethyleneguanidine phosphate;

QA: MIC value (ppm) of 3-isothiazolone out of a mixture;

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QB: MIC value (ppm) of polyhexamethyleneguanidine out of a mixture.

As represented in the above Table 1, it can be seen that the same microorganism killing effect (SI was 0.53 in EXAMPLE 1) can be obtained even though only a half amount of 3-isothiazolone is used and the amount of polyhexamethyleneguanidine phosphate used is reduced to 9.8 ppm. The same microorganism killing effect (SI was 0.63 in EXAMPLE 6) can also be obtained even though only a half amount of polyhexamethyleneguanidine phosphate is used and only 2.4 ppm of 3-isothiazolone is used. Therefore, it can be shown that a mixture of the above two compounds can inhibit the growth of bacteria 15 more effectively than the single use of each of the two compounds, and the synergistic action ratio of 3-isothiazolone and polyhexamethyleneguanidine phosphate is most preferable in the range of 1:1 to 1:4, where SI is from 0.53 to 0.63.

[TEST EXAMPLE 1 AND COMPARATIVE EXAMPLE 1]

The minimum inhibition concentration (MIC) values of a biocide composition in which 3-isothiazolone and polyhexamethyleneguanidine phosphate are mixed in a ratio of 1:4 (1 wt% of 3-isothiazolone and 15 wt% of 25 wt% poly-hexamethyleneguanidine phosphate) and of a single biocide of 1.5 wt% 3-isothiazolone were measured on 7 types of individual strains. After 25 diluting a biocide using 96 multi wall plates according to the two fold continuous dilution method, microorganisms were inoculated at a concentration of 104 CFU/ml. Then, after culturing at 30 degrees centigrade for 48 hours, the MIC values were measured by observation with the naked eye of the growth of microorganisms on the basis of muddiness. The results are represented in

Table 2.

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The muddiness was observed using Tryptic Soy Broth manufactured by Difco Corporation as a medium in order to measure MIC values and strains of Enterobacter aerogens ATCC 13048, Staphylococcus aureus ATCC 6538, epidermis ATCC 155, Bacillus subtilis ATCC 6984. 5 Staphylococcus Saccharomyces cerevisiae ATCC 9763, Rhizopus oryzae ATCC 10404, Aspergillus niger ATCC 9642 were used from among the strains which were used in EXAMPLE 1.

[Table 2] MIC test results on microorganisms of a biocide mixture and a single biocide of 1.5 wt% 3-isothiazolone (units: ppm):

Strains Used	COMPARATIVE EXAMPLE	TEST EXAMPLE
	(1.5% isothiazolone)	(Biocide mixture)
Enterobacter aerogens ATCC 13048	390	195
Staphylococcus aureus ATCC 6538	195	195
Staphylococcus epidermis ATCC 155	390	97
Bacillus subtilis ATCC 6984	390	195
Saccharomyces cerevisiae ATCC 9763	390	390
Rhizopus oryzae ATCC 10404	390	195
Aspergillus niger ATCC 9642	195	195

As can be shown from the above Table 2, a biocide mixture in which 3-15 isothiazolone and 25 wt% polyhexamethyleneguanidine phosphate are mixed using the synergistic index is much more effective in controlling microorganisms than is a biocide with a single component of 1.5 wt% 3-isothiazolone.

ITEST EXAMPLE 2 AND COMPARATIVE EXAMPLE 2]

In order to determine the killing time to kill microorganisms when the biocide mixture used in TEST EXAMPLE 1 and 1.5% 3-isothiazolone that was used in COMPARATIVE EXAMPLE 1, measurements were made of the immediate effectiveness, the durability, and the number of strains by taking a strain solution at times corresponding to 0 hours, 3 hours, 24 hours, 48 hours, 25 72 hours, and 96 hours after respectively putting in 50, 100, and 200 ppm of a biocide mixture and 1.5% 3-isothiazolone into the cooling water of a polymerization cooling tower having a microorganism concentration of about 104 CFU/ml..., A solution in which a biocide was not added was used as a blank. The measured results on the strain reduction ratios according to the time when the biocide mixture in which 3-isothiazolone and polyhexamethyleneguanidine phosphate was mixed are presented using a synergistic index, as well as data for single component biocide of 1.5 wt% 3-isothiazolone, are represented in Table 3 and Table 4.

[Table 3]

Measured results on the reduction ratio of microorganisms according to the concentration of a biocide mixture verses time (units: CFU/ml):

_	_
1	n
1	v

		0	3 hours	24 hours	48 hours	72 hours	96 hours
Bli	ank	13,000	> 13,000	> 13,000	> 13,000	> 13,000	> 13,000
Biocide	50 ppm	13,000	210	140	90	40	50
mixture	100 ppm	13,000	80	170	90	50	30
	200 ppm	13,000	50	90	50	40	50

[Table 4]

Measured results on the reduction ratio of microorganisms according to

the concentration verses time of 1.5 wt% of 3-isothiazolone (units: CFU/ml):

		0	3 hours	24 hours	48 hours	72 hours	96 hours
Bla	nk	13,000	> 13,000	> 13,000	> 13,000	> 13,000	> 13,000
1.5 wt% of	50 ppm	13,000	12,000	450	> 10,000	> 10,000	> 10,000
isothiazolon	100 ppm	13,000	11,400	370	6,250	> 10,000	> 10,000
e	200 ppm	13,000	5,300	200	2,100	> 10,000	> 10,000

As can be shown in the above Table 3 and Table 4, the single compound 1.5 wt% of 3-isothiazolone biocide shows sterilizing capability after 24 hours, while the immediate effectiveness is low and there is also a secondary 20 propagation of microorganisms after 48 hours. The biocide mixture of 3isothiazolone and polyhexamethyleneguanidine phosphate achieves synergistic effects as seen in immediate effectiveness, durability, and superior sterilizing effects on microorganisms as compared to the single component biocide. 3-isothiazolone and when that shown Therefore, it can be polyhexamethyleneguanidine phosphate are mixed, the disadvantage of 3problems of and the immediate effectiveness with isothiazolone

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polyhexamethyleneguanidine phosphate associated with antibiotic spectrum are mutually compensated, and sterilizing capability is correlated with a synergistic effect index.

As described in the above, a biocide composition of the present invention can be effectively used in the wide control of microorganisms in various industrial fields such as water treatment, disinfectants, etc., since it has immediate effectiveness and durability, as well as applicability to a wide antibiotic spectrum. Furthermore, a biocide composition of the present invention can effectively control industrial water microorganism contamination and the actual living environment of the microorganisms since it is more effective in controlling the microorganisms and has greater immediate sterilizing capability than does a single compound biocide. A biocide composition of the present invention has an effect to lower the emergence frequency of a resistant strain as compared a single compound biocide, achieving this using a mixture of biocides having the different working mechanism.

While the present invention has been described in detail with reference to the preferred embodiments, those skilled in the art will appreciate that various modifications and substitutions can be made thereto without departing from the spirit and scope of the present invention as set forth in the appended claims.

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WHAT IS CLAIMED IS:

A biocide composition comprising 3-isothiazolone of the following General Formula 1 and polyhexamethyleneguanidine phosphate of the following General Formula 2:

[General Formula 1]

[General Formula 2]

$$\begin{array}{c} \text{NH} \\ \parallel \\ -\{(\text{CH}_2)_6 - \text{NH} - \text{C} - \text{NH} - \frac{1}{m} \cdot \text{nH}_3 \text{PO}_4 \end{array}$$

where R is hydrogen or chlorine in the above General Formula 1, m is an integer from 4 to 7 and n is an integer from 1 to 14 in the above General Formula 2.

- 2. A biocide composition in accordance with claim 1 wherein, the mixing ratio of the above 3-isothiazolone and polyhexamethyleneguanidine phosphate is a weight ratio of 1:1 to 1:65.
- 3. A biocide composition in accordance with claim 1 wherein, the above 3-isothiazolone is a mixture in which 3-isothiazolone having R of hydrogen and 3-isothiazolone having R of chlorine are mixed in a weight ratio of 1:20 to 20:1 and the mixing ratio of the above 3-isothiazolone and polyhexamethyleneguanidine phosphate is from 1:1 to 1:4.
- 4. A biocide composition in accordance with claim 1 characterized in that before the use of the above biocide composition, it is mixed with a media selected from the group consisting of cooling water of an industrial process, disinfectant, paint, antiseptic for latex, additives for cosmetics, additives for emulsion products, slime control chemicals for textile weaving, paper slime

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control agent, antiseptic for leather goods, and antiseptic for metal processing oil.

5. A sterilizing method of killing or restraining the growth of bacteria, fungi and/or algae by putting a biocide composition of claim 1 into the area that is contaminated by bacteria, fungi and/or algae.

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INTERNATIONAL SEARCH REPORT

International application No. PCT/KR99/00687

A. CLASSIFICATION OF SUBJECT MATTER		
IPC ⁷ : A 01 N 43/80, 47/44		
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Date of the actual completion of the international search 1 February 2000 (01.02.00)	29 March 2000 (29.03.)	
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INTERNATIONAL SEARCH REPORT

Information on patent family members

International application No. PCT/KR 99/00687

JP A2 10175809 30-06-1998 none	Patent document cited in search report	Publication date	Patent family member(s)	Publication date
	JP A2 10175809	30-06-1998	none	
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REQUEST

The undersigned requests that the present

international application be processed according to the Patent Cooperation Treaty.	Name of receiving Office and "PCT International Application"				
	Applicant's or agent's file reference (if desired) (12 characters maximum) OPP990727KR				
Box No. I TITLE OF INVENTION BIOCIDE C	COMPOSITION AND STERILIZATION METHOD				
USING THE SAME					
Box No. II APPLICANT					
Name and address: (Family name followed by given name; for designation. The address must include postal code and name of a address indicated in this Box is the applicant's State (that is, coun of residence is indicated below.)	r a legal entity, full official country. The country of the http://doi.org/10.1001/10.	,			
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Sheet	No.		.2

	ND/OR (FURTHER) I	NVENTOR(S)
If none of the following sub-boxes is used, th	nis sheet should not be in	cluded in the request.
Name and address: (Family name followed by given name; for a leading address: (Family name followed by given name; for a leading address indicated in this Box is the applicant's State (that is, country) of residence is indicated below.)	legal entity, full official ntry. The country of the ofresidence if no State	This person is:
KIM, Jin-Man		applicant only .
Cheonrok Apt. 3-306, Yuljeon-dong, Changan-l	ku	X applicant and inventor
Suwon-city, Kyungki-do, 440-320		inventor only (If this check-box
Republic of Korea		is marked, do not fill in below.)
State (that is, country) of nationality: KR	State (that is, country)	of residence: KR
	ites of America	e United States America only the States indicated in the Supplemental Box
Name and address: (Family name followed by given name: for a le designation. The address must include postal code and name of coun address indicated in this Box is the applicant's State (that is, country) of residence is indicated below.)	egal entity, full official try. The country of the of residence if no State	This person is:
PARK, Jeong-Ho		
Dongshin Apt. 201-1109, Jungja 1-dong, Chang	gan-ku	X applicant and inventor
Suwon-city, Kyungki-do, 440-301	į	inventor only (If this check-box is marked, do not fill in below.)
Republic of Korea	<u> </u>	
State (that is, country) of nationality: KR	State (that is, country) o	f residence: KR
This person is applicant for the purposes of: all designated States all designated States.	States except	United States the States indicated in the Supplemental Box
Name and address: (Family name followed by given name: for a legal designation. The address must include postal code and name of count address indicated in this Box is the applicant's State (that is, country) of residence is indicated below.)	gal entity, full official iry. The country of the of residence if no State	This person is:
CHO, Myung-Ho		applicant only
Samsung Apt. 1-805, Kwonseon-dong		X applicant and inventor
Kwonseon-ku, Suwon-city, Kyungki-do, 441-39	90	inventor only (If this check-box is marked, do not fill in below.)
Republic of Korea	l	is marked, do not jui in below,
State (that is, country) of nationality: KR	State (that is, country) o	f residence: KR
This person is applicant for the purposes of: all designated States all designated the United State		United States America only the States indicated in the Supplemental Box
Name and address: (Family name followed by given name; for a leg designation. The address must include postal code and name of count address indicated in this Box is the applicant's State (that is, country) of residence is indicated below.)	gal entity, full official try. The country of the of residence if no State	This person is:
HAHN, Soon-Jong		applicant only
Kwanak-Hyundai Apt. 123-1402, Bongcheon 3-	dong	X applicant and inventor
Kwanak-ku, Seoul, 151-053		inventor only (If this check-box is marked, do not fill in below.)
Republic of Korea		is marked, do not fill the below.y
State (that is, country) of nationality: KR	State (that is, country) of	residence: KR
This person is applicant all designated States all designated States all designated States	States except X the es of America	United States the States indicated in the Supplemental Box
Further applicants and/or (further) inventors are indicated on	another continuation shee	et.

Box	No.V	DESIGNATION OF ST. ES			
The	follov	ving designations a reby made under Rule 4.9)(:1)	mark	the applicat
Regio	onal F	Patent	(4)	urn	the applicable seck-boxes, at least one must be marked):
		211 Zimbabwe, and any other State which is a Conf	racui	ng Stu	tho, MW Malawi, SD Sudan, SZ Swaziland, UG Uganda, te of the Harare Protocol and of the PCT
	ΕA	Eurasian Patent: AM Armenia AZ Azerbaijan	RY	Relai	rus, KG Kyrgyzstan, KZ Kazakhstan, MD Republic of inistan, and any other State which is a Contracting State
Ø	EP	European Patent: AT Austria, BE Belgium, CH : DK Denmark, ES Spain, FI Finland, FR France, GB	Unite	የለ ዜ ነና	ritzerland and Liechtenstein, CY Cyprus, DE Germany, agdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, by other State which is a Contracting State of the European
	OA	any other State which is a member State of OAPI an	da C	lontra Contra	n Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, uritania, NE Niger, SN Senegal, TD Chad, TG Togo, and cting State of the PCT (if other kind of protection or treatment
Natio	nal P	atent (if other kind of protection or treatment desired, special	 	 	
רח	AT	A Ib	_		·
		Albania		LS	Lesotho
LJ		Armenia		LT	Lithuania
		Austria		LU	Luxembourg
X	ΑU	Australia			Latvia
	ΑZ	Azerbaijan	$\overline{\Box}$	MD	Republic of Moldova
		Bosnia and Herzegovina			Madagascar
n		Barbados	=		-
		Bulgaria		IVL	The former Yugoslav Republic of Macedonia
H			_		••••••
=		Brazil		MN	Mongolia
		Belarus		MW	/ Malawi
X		Canada		MX	Mexico
□.		and LI Switzerland and Liechtenstein		NO	Norway
X	CN	China		NZ	New Zealand
	CU.	Cuba			Poland
		Czech Republic	$\overline{\Box}$		Portugal
		Germany	\Box		Romania
Ē		Denmark			
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	ES	Spain		SD	Sudan
				SE	Sweden
	FI	Finland		SG	Singapore
		United Kingdom		SI	Slovenia
Ш		Grenada		SK	Slovakia
		Georgia		SL	Sierra Leone
	GH	Ghana		TJ	Tajikistan
	GM	Gambia		TM	Turkmenistan
	HR	Croatia	ī	TR	Turkey
		Hungary	$\overline{\Box}$	TT	Trinidad and Tobago
$\overline{\Box}$	ID	Indonesia			
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	IS	Iceland	_		• • • • • • • • • • • • • • • • • • • •
\mathbf{x}		Japan		UZ	Uzbekistan
		Kenya		VN	Viet Nam
	KG	Kyrgyzstan		YU	Yugoslavia
	KP	Democratic People's Republic of Korea			Zimbabwe
			Che		xes reserved for designating States (for the purposes of patent) which have become party to the PCT after
			issu	nonal	f this sheet:
닏		Kazakhstan			
		Saint Lucia			•••••
	LK	Sri Lanka			••••••
	LR	Liberia .			•••••
Precau	tiona	ry Designation Statement: In addition to the designat	ions		above, the applicant also makes under Rule 4.9(b) all other

Precautionary Designation Statement: In addition to the designations made above, the applicant also makes under Rule 4.9(b) all other designations which would be permitted under the PCT except any designation(s) indicated in the Supplemental Box as being excluded from the scope of this statement. The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation of a designation consists of the filing of a notice specifying that designation and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.)

Sheet No. .4....

Box No. VI PRIORITY C	I AVM		Further	priority claims are indicated	d in the Cumplemental Day		
Filing date	Number		Further priority claims are indicated in the Supplemental Box. Where earlier application is:				
of earlier application (day/month/year)	of earlier applic	ation	national application country		international application: receiving Office		
item(1) 16 November, 1998 (16/11/1998)	98-49095		KR				
item (2)							
item (3)							
The receiving Office is required of the earlier application(single purposes of the present into) (only if the earlie	r applic	cation was filed with th	he Office which for the			
* Where the earlier application is a Convention for the Protection of In-	an ARIPO application dustrial Property for	n, it is m which th	andatory to indicate in th hat earlier application wa:	e Supplemental Box at least o s filed (Rule 4.10(b)(ii)). See .	ne country party to the Paris Supplemental Box.		
	NAL SEARCHIN						
Choice of International Search (if two or more International Sear competent to carry out the internat the Authority chosen: the two-letter	rching Authorities at tional search, indicat	e sear	quest to use results of e ch has been carried out by c (day/month/year)	arlier search; reference to requested from the International Number	tional Searching Authority):		
ISA / AT	tout may be assumed.		o (uay/monuoyea/)	Number	Country (or regional Office)		
Box No. VIII CHECK LIST;	; LANGUAGE O	F FILI	NG				
This international application co	:			anied by the item(s) marke	d below:		
request : 4	1. 🐹 1ec		ation sheet				
description (excluding	-		igned power of attorney				
sequence listing part) : 11	1 =	-		; reference number, if any	:		
claims : 2	1		explaining lack of signs				
abstract : 1				Box No. VI as item(s):			
drawings :			of international application				
sequence listing part of description :				eposited microorganism or			
Total number of shares 10	1			sence listing in computer re	adable form		
Figure of the drawings which	3. L 0a		iguage of filing of the				
should accompany the abstract:		inte	mational application:	English			
	F APPLICANT O				-		
Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request). KIM, WON-HO							
		For rec	ceiving Office use only				
Date of actual receipt of the p international application:	ourported				2. Drawings:		
 Corrected date of actual receitimely received papers or draw the purported international ap 	wings completing				received:		
 Date of timely receipt of the r corrections under PCT Article 	required e 11(2):				not received:		
5. International Searching Author (if two or more are competent	ority): ISA /		6. Transmit until sea	rtal of search copy delayed rch fee is paid.			
Date of receipt of the record copy by the International Bureau:		r Intern	ational Exceau use only				

rcı	For receiving Office use only
FEE CALCULATION SHEET	
Annex to the Request	International application No.
Applicant's or agent's file reference OPP990727KR	Date stamp of the receiving Office
Applicant SK Chemicals, et al.	
CALCULATION OF PRESCRIBED FEES	
1. TRANSMITTAL FEE	W 45,000 T
2. SEARCH FEE International search to be carried out by <u>Austrian Paten</u> (If two or more International Searching Authorities are competent in relation application, indicate the name of the Authority which is chosen to carry out the international searching Authority which is chosen to carry out the international search of the Authority which is chosen to carry out the international search of the Authority which is chosen to carry out the international search of the search	t Office on to the international international search.)
3. INTERNATIONAL FEE	
Basic Fee The international application contains 18 sheets.	
first 30 sheets	OO [b1]
x =	b2
remaining sheets additional amount	
Add amounts entered at b1 and b2 and enter total at B	W 517,100 B
Designation Fees The international application contains6 designations.	
	N 715,800 D
number of designation fees amount of designation fee payable (maximum 11)	
Add amounts entered at B and D and enter total at I	W 1,232,900 I
4. FEE FOR PRIORITY DOCUMENT (if applicable)	P P
5. TOTAL FEES PAYABLE	W 1,481,900 TOTAL
The designation fees are not paid at this time.	
MODE OF PAYMENT	
authorization to charge deposit account (see below) cheque X cash postal money order revenue stamps	coupons other (specify):
DEPOSIT ACCOUNT AUTHORIZATION (this mode of payment m.	ay not be available at all receiving Offices)
The RO/ is hereby authorized to charge the total fees i	
is hereby authorized to charge any deficiency deposit account.	y or credit any overpayment in the total fees indicated above to my
is hereby authorized to charge the fee for prep Bureau of WIPO to my deposit account.	paration and transmittal of the priority document to the International
Deposit Account No. Date (day/month/year)	Signature

PATENT COOPERATION T

From the RECEIVING OFFICE

Facsimile No. 82-42-472-3466 Form PCT/RO/105 (July 1992) ТV

To: KIM, Won Ho			PO	CT		
702, Teheran Bldg., 825-33 Yoksam-dong, Kangnam-ku, Seoul 135-080, Republic of KOREA		NOTIFICATION OF THE INTERNATIONAL APPLICATION NUMBER AND OF THE INTERNATIONAL FILING DATE				
			(PCT Ru	le 20.5(C))		
		Date of mailing (day/month/year)	19 NOVEMI	BER 1999 (19.11.1999)		
Applicant's or agent's file reference OPP990727KR		IMP	ORTANT NO	TIFICATION		
International application No.	International filing date	(day/month/year)	Priority date	(day/month/year)		
PCT/KR99/00687	16 NOVEMBER 19	999 (16.11.1999)	16 NOVEM	1BER 1998 (16.11.1998)		
Applicant SK CHEMICALS et al Title of the invention BIOCIDE COMPOSITION AND S	TERILIZATION ME	THOD USING TH	E SAME	YOU ME NOV. 2 4. 1999 FRECEIVED		
1. The applicant is hereby notified that the international filing date indicated 2. The applicant is further notified that was transmitted to the International Market State Stat	above. t the record copy of the ational Bureau on d to the International Bute the International Bureau on the International Bureau on Bureau on the International Bureau on the International Security clean	ne international applic sureau for the reason au*:	ation:			
÷	·	•				
* The International Bureau monitors to (with Form PCT/IB/301) of its receif the priority date, the International B	pt. Should the record co	ppy not have been reco	eived by the ex	d will notify the applicant piration of 14 months from		
Name and mailing address of the receiving	g Office	Authorized officer				
Korean Industrial Property Office Government Complex-Taejon, Dunsan-o Metropolitan City 302-701, Republic of	long, So-ku, Tacion	COMMISSIO	NER	信出扈		
Facsimile No. 82-42-472-3466	120100	Telephone No. 82-4	42-481-5213			

From the INTERNATIONAL SEARCHING AUTHORITY

To:

KIM, Won Ho 702, Teheran Bd., 825-33, Yoksam-dong, Kangnam-ku Seoul 135-080 Republic of Korea



NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL SEARCH REPORT OR THE DECLARATION

RECEIVED	(PCT Rule 44.1)					
S 1 PSCO	Date of mailing (day/month/year) 29 Mrz. 2000 (29.03.00)					
Applicant's or agent's file reference OPP990727KR	FOR FURTHER ACTION See paragraphs 1 and 4 below					
International application No. PCT / KR 99/00687	International filing date (day/month/year) 16 Nov. 1999 (16.11.99)					
Applicant	L					
SK CHEMICALS et al.						
1. X The applicant is hereby notified that the international s	earch report has been established and is transmitted herewith.					
Filing of amendments and statement under Article 19: The applicant is entitled, if he so wishes, to amend the claims of the international application (see Rule 46):						
When? The time limit for filing such amendme international search report; however, for r	nts is normally two months from the date of transmittal of the nore details, see the notes on the accompanying sheet.					
Where? Directly to the International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35						
For more detailed instructions, see the notes on the accompanying sheet.						
The applicant is hereby notified that no international sea 17(2)(a) to that effect is transmitted herewith.	rch report will be established and that the declaration under Article					
3. With regard to the protest against payment of (an) ad	Iditional fee(s) under Rule 40.2, the applicant is notified that:					
the protest together with the decision thereon h applicant's request to forward the texts of both to	as been transmitted to the International Bureau together with the he protest and the decision thereon to the designated Offices.					
no decision has been made yet on the protest; the	ne applicant will be notified as soon as a decision is made.					
4. Further action(s): The applicant is reminded of the follo	wing:					
applicant wishes to avoid or postpone publication, a notic	al application will be published by the International Bureau. If the see of withdrawal of the international application, or of the priority Rules 90bis.1 and 90bis.3, respectively, before the completion of					
Within 19 months from the priority date, a demand for inte wishes to postpone the entry into the national phase until	Within 19 months from the priority date, a demand for international preliminary examination must be filed if the applicant wishes to postpone the entry into the national phase until 30 months from the priority date (in some Offices even later).					
Within 20 months from the priority date, the applicant must pall designated Offices which have not been elected in the door could not be elected because they are not bound by Chapter 1.	perform the prescribed acts for entry into the national phase before emand or in a later election within 19 months from the priority date apter II.					
Name and mailing address of the ISA/	Authorized officer					
AUSTRIAN PATENT OFFICE Kohlmarkt 8-10	Koch					
Facsimile No. A-1014 Vienna +43/1/534.24 - 200	+43 / 1 / 534 24 - 450 Telephone No.					

+43 / 1 / 534 24 - 200

NOTES TO FORM PCT/ISA/220

These Notes are intended to give the basic instructions concerning the filing of amendments under Article 19. The Notes are based on the requirements of the Patent Cooperation Treaty, the Regulations and the Administrative Instructions under that Treaty. In case of discrepancy between these Notes and those requirements, the latter are applicable. For more detailed information, see also the PCT Applicant's Guide, a publication of WIPO.

In these Notes, "Article," "Rule" and "Section" refer to the provisions of the PCT, the PCT Regulations and the PCT Administrative Instructions, respectively.

INSTRUCTIONS CONCERNING AMENDMENTS UNDER ARTICLE 19

The applicant has, after having received the international search report, one opportunity to amend the claims of the international application. It should however be emphasized that, since all parts of the international application (claims, description and drawings) may be amended during the international preliminary examination procedure, there is usually no need to file amendments of the claims under Article 19 except where, e.g. the applicant wants the latter to be published for the purposes of provisional protection or has another reason for amending the claims before international publication. Furthermore, it should be emphasized that provisional protection is available in some States only.

What parts of the international application may be amended?

Under Article 19, only the claims may be amended.

During the international phase, the claims may also be amended (or further amended) under Article 34 before the International Preliminary Examining Authority. The description and drawings may only be amended under Article 34 before the International Preliminary Examining Authority.

Upon entry into the national phase, all parts of the international application may be amended under Article 28 or, where applicable, Article 41.

When? Within 2 months from the date of transmittal of the international search report or 16 months from the priority date, whichever time limit expires later. It should be noted, however, that the amendments will be considered as having been received on time if they are received by the International Bureau after the expiration of the applicable time limit but before the completion of the technical preparations for international publication (Rule 46.1).

Where not to file the amendments?

The amendments may only be filed with the International Bureau and not with the receiving Office or the International Searching Authority (Rule 46.2).

Where a demand for international preliminary examination has been/is filed, see below.

How? Either by cancelling one or more entire claims, by adding one or more new claims or by amending the text of one or more of the claims as filed.

A replacement sheet must be submitted for each sheet of the claims which, on account of an amendment or amendments, differs from the sheet originally filed.

All the claims appearing on a replacement sheet must be numbered in Arabic numerals. Where a claim is cancelled, no renumbering of the other claims is required. In all cases where claims are renumbered, they must be renumbered consecutively (Administrative Instructions, Section 205(b)).

The amendments must be made in the language in which the international application is to be published.

What documents must/may accompany the amendments?

Letter (Section 205(b)):

The amendments must be submitted with a letter.

The letter will not be published with the international application and the amended claims. It should not be confused with the "Statement under Article 19(1)" (see below, under "Statement under Article 19(1)").

The letter must be in English or French, at the choice of the applicant. However, if the language of the international application is English, the letter must be in English; if the language of the international application is French, the letter must be in French.



The letter must indicate the differences between the claims as filed and the claims as amended. It must, in particular, indicate, in connection with each claim appearing in the international application (it being understood that identical indications concerning several claims may be grouped), whether

- (i) the claim is unchanged;
- (ii) the claim is cancelled;
- (iii) the claim is new;
- (iv) the claim replaces one or more claims as filed:
- (v) the claim is the result of the division of a claim as filed.

The following examples illustrate the manner in which amendments must be explained in the accompanying letter:

- 1. [Where originally there were 48 claims and after amendment of some claims there are 51]: "Claims 1 to 29, 31, 32, 34, 35, 37 to 48 replaced by amended claims bearing the same numbers; claims 30, 33 and 36 unchanged; new claims 49 to 51 added."
- [Where originally there were 15 claims and after amendment of all claims there are 11]: "Claims 1 to 15 replaced by amended claims 1 to 11."
- [Where originally there were 14 claims and the amendments consist in cancelling some claims and in adding new claims]:
 - "Claims 1 to 6 and 14 unchanged; claims 7 to 13 cancelled; new claims 15, 16 and 17 added." or "Claims 7 to 13 cancelled; new claims 15, 16 and 17 added; all other claims unchanged."
- 4. [Where various kinds of amendments are made]: "Claims 1-10 unchanged; claims 11 to 13, 18 and 19 cancelled; claims 14, 15 and 16 replaced by amended claim 14; claim 17 subdivided into amended claims 15, 16 and 17; new claims 20 and 21 added."

"Statement under Article 19(1)" (Rule 46.4)

The amendments may be accompanied by a statement explaining the amendments and indicating any impact that such amendments might have on the description and the drawings (which cannot be amended under Article 19(1)).

The statement will be published with the international application and the amended claims.

It must be in the language in which the international application is to be published.

It must be brief, not exceeding 500 words if in English or if translated into English.

It should not be confused with and does not replace the letter indicating the differences between the claims as filed and as amended. It must be filed on a separate sheet and must be identified as such by a heading, preferably by using the words "Statement under Article 19(1)."

It may not contain any disparaging comments on the international search report or the relevance of citations contained in that report. Reference to citations, relevant to a given claim, contained in the international search report may be made only in connection with an amendment of that claim.

Consequence if a demand for international preliminary examination has already been filed

If, at the time of filing any amendments and any accompanying statement, under Article 19, a demand for international preliminary examination has already been submitted, the applicant must preferably, at the time of filing the amendments (and any statement) with the International Bureau, also file with the International Preliminary Examining Authority a copy of such amendments (and of any statement) and, where required, a translation of such amendments for the procedure before that Authority (see Rules 55.3(a) and 62.2, first sentence). For further information, see the Notes to the demand form (PCT/IPEA/401).

Consequence with regard to translation of the international application for entry into the national phase

The applicant's attention is drawn to the fact that, upon entry into the national phase, a translation of the claims as amended under Article 19 may have to be furnished to the designated/elected Offices, instead of, or in addition to, the translation of the claims as filed.

For further details on the requirements of each designated/elected Office, see the PCT Applicant's Guide, Volume II.



PCT

INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

ti di reference		
Applicant's or agent's file reference OPP990727KR	FOR FURTHER see Notification of ACTION (Form PCT/ISA/22	Transmittal of International Search Report 0) as well as, where applicable, item 5 below.
	International filing date (day/month/year)	(Earliest) Priority Date (day/month/year)
International application No.	16 November 1999 (16.11.99)	16 November 1998 (16.11.98)
PCT/KR 99/00687	16 November 1999 (16.11.99)	
Applicant		
SK CHEMICALS et al.		
- Land has been	premared by this International Searching A	Authority and is transmitted to the applicant
according to Article 18. A copy is being t	ransmitted to the International Bureau.	
This international search report consists of		
1	by a copy of each prior art document cite	d in this report.
language in which it was filed,	, unless otherwise indicated under this iter	the basis of the international application in the m.
the international search wa	s carried out on the basis of a translation of	of the international application furnished to this
b. With regard to any nucleotide search was carried out on the	e and/or amino acid sequence disclosed in basis of the sequence listing:	in the international application, the international
contained in the internation	nal application in written form.	·
filed together with the into	ernational application in computer readab	le form.
furnished subsequently to	this Authority in written form.	
furnished subsequently to	this Authority in computer readable form	. .
international application	as filed has been furnished.	ng does not go beyond the disclosure in the
the statement that the info	ormation recorded in computer readable for	orm is identical to the written sequence listing has
2. Certain claims were fou	nd unsearchable (See Box I).	
3. Unity of invention is lack		
4. With regard to the title,		
the text is approved as su	bmitted by the applicant.	
	hed by this Authority to read as follows:	• .
5. With regard to the abstract,		
the text is approved as su	bmitted by the applicant.	
	had according to Pule 38 2(h) by this At	athority as it appears in Box III. The applicant may, arch report, submit comments to this Authority.
1	published with the abstract is Figure No.:	
as suggested by the appli		None of the figures.
because the applicant fai		
	r characterizes the invention.	

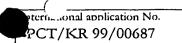
INTERNATION...L SEARCH REPORT

_	
	International application No.
	PCT/KR99/00687

A. CLAS	SIFICATION OF SUBJECT MATTER	101/1203/100					
1	01 N 43/80, 47/44						
	According to International Patent Classification (IPC) or to both national classification and IPC B. FIELDS SEARCHED						
	ocumentation searched (classification system followe	d by classification symbols)					
IPC ⁷ : A0	-	a by blassification symbols)					
IPC. Au	JIN						
Documentati	Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched						
Electronic da	ta base consulted during the international search (na	me of data base and, where practicable, search	ch terms used)				
WPI							
C. DOCU	MENTS CONSIDERED TO BE RELEVANT						
Category*	Citation of document, with indication, where appropriate appropria	priate, of the relevant passages	Relevant to claim No.				
A DATABASE WPI ON EPOQUE, week199836, London: 1-5 Derwent Publications Ltd., AN 1998-422258, class A97, JP 10175809 A (NAGASE KASEI KOGYO KK), abstract.							
		·					
i							
			- 1 				
Further of	documents are listed in the continuation of Box C.	See patent family annex.					
	egories of cited documents:	"T" later document published after the internation	onal filing date or priority				
	defining the general state of the art which is not to be of particular relevance	date and not in conflict with the application the principle or theory underlying the inven					
.E" earlier appl	ication or patent but published on or after the international	"X" document of particular relevance; the claim	ed invention cannot be				
tiling date L" document v	which may throw doubts on priority claim(s) or which is	considered novel or cannot be considered to when the document is taken alone	involve an inventive step				
cited to esta	ablish the publication date of another citation or other	"Y" document of particular relevance; the claim					
	on (as specified) eferring to an oral disclosure, use, exhibition or other	considered to involve an inventive step who combined with one or more other such doc					
means		being obvious to a person skilled in the art					
	ublished prior to the international filing date but later than date claimed	"&" document member of the same patent famil	у				
	tual completion of the international search	Date of mailing of the international search	report				
l Februa	ary 2000 (01.02.00)	29 March 2000 (29.03.00)				
Name and mai	lling adress of the ISA/AT	Authorized officer					
	atent Office						
Kohlmarkt	8-10; A-1014 Vienna	SCHNASS					
Facsimile No.	1/53424/535	Telephone No. 1/53424/217					

Form PCT/ISA/210 (second sheet) (July 1998)

INTERNAT NAL SEARCH REPORT Information patent family members



Pate ii	ent docum int cited n search r port	Publication date	Patent family member(s)	Publication date
JP A2		30-06-1998	none	
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[별지 제55호서식]

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To : Commi the Ko		ial Property (Office					
International Application No. PCT/KR99/00687								
Amaliaant	Name	SK Chemic	als et al.	Resider Reg. N			Country o	I KK
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Contents of	Former	SK Chemicals et al.						
Notification	New	SK Chemicals Co., Ltd., et al.						
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	Date(day/month/ye	ear)					
09/05/2001 Applicant (Agent) KIM, Won-Ho (Se								
** Attached Document(s): * Attached Document(s): ** Attached Document(s): ** Attach								
	1. A copy of the document(s) substantiating the contents of notification							
2. A copy of the document(s) substantiating the power of attorney, if any								

50285-10421민 99. 6. 2. 개정승인 210mm×297mm (보존용지(2종) 70g/m²)